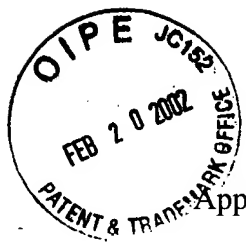


1# 86



PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Application of

Applicant : Stephen N. Vaughn  
Serial No. : 08/943,399  
Filed : 10/3/97  
Title : Method for Increasing Light Olefin Yield by Conversion of a  
Heavy Hydrocarbon Fraction of a Product to Light Olefins  
Docket : 97B058/3  
Examiner : Walter D. Griffin  
Group Art Unit : 1764

DECLARATION UNDER 37 C.F.R. § 1.132

Honorable Commissioner for Patents  
Washington, D.C. 20231

Dear Sir:

RECEIVED  
FEB 26 2002  
TC 1700

I, STEPHEN N. VAUGHN, hereby declare as follows:

1. I reside at 1111 Southern Hills Rd., Kingwood, Texas 77339.
2. I am the only named inventor on U.S. Patent Application Serial No. 08/943,399 (the '399 application) titled "Method for Increasing Light Olefin Yield by Conversion of a Heavy Hydrocarbon Fraction of a Product to Light Olefins", filed on October 3, 1997.
3. I received a B.S. in Nuclear Engineering in 1974 from Kansas State University and a Ph.D. in Engineering in 1980 from Kansas State University.
4. I have worked in the catalysis field for approximately 21 years and in the field of methanol to olefin catalysis since 1993.

5. I have reviewed the '399 application, the Office Action dated June 26, 2001, and the references relied upon therein including U.S. Patent No. 4,527,001 to Kaiser (the Kaiser patent).

6. The Kaiser patent is directed to olefin interconversion including the conversion of butene over a SAPO catalyst to ethylene and propylene. There is no teaching of conversion of methanol to olefins. Likewise, there is no teaching of the conversion of a reaction feed comprising methanol and butene. The first column of Table III in the Kaiser patent indicates that when 1-butene was subjected to reaction conditions over a SAPO catalyst, the product included 11.77% propylene and only 1.78% ethylene. The weight ratios of ethylene to propylene produced according to the data shown in the three columns of Table III are 0.10, 0.12 and 0.13. Similarly, the first column of Table VIII of the Kaiser patent indicates that when 1-butene was subjected to reaction conditions over a SAPO catalyst, the product included 6.47% propylene and only 2.18% ethylene. The weight ratios of ethylene to propylene produced according to the data shown in the three columns of Table VIII are 0.22, 0.34 and 0.26. Since all of these ratios are less than 1, the Kaiser patent indicates that in the conversion reaction of 1-butene, propylene is highly favored over ethylene.

7. The Table in Example 1 of the '399 Application indicates, in relevant part:

**Conversion and Selectivities of 1-Butene, MeOH, and MeOH + 1-Butene Reactions  
(Reproduced From The '399 Application, Example 1)**

Feed	Conversion	C <sub>2</sub> <sup>=</sup>	C <sub>3</sub> <sup>=</sup>	C <sub>5</sub> <sup>+</sup>
1-butene	34.23%	10.57%	66.29%	21.74%
MeOH	100.00%	51.06%	34.19%	2.08%
MeOH + 1-butene	84.32%	45.72%	41.46%	2.67%

8. My experimental data disclosed in the second row of the Table in Example 1 of the '399 Application, and reproduced above, indicates that the MeOH conversion

reaction in the presence of SAPO-34 catalyst produces 51.06% ethylene and 34.19% propylene. Thus, ethylene is favored over propylene in the MeOH conversion reaction over SAPO-34, which is a small pore non-zeolitic molecular sieve catalyst.

9. The *actual* yields of the component reactions and combined reaction may be calculated by multiplying the respective conversions by the selectivities, as follows:

**Actual Yields of 1-Butene, MeOH, and MeOH + 1-Butene Reactions  
(Calculated by Multiplying Conversion Times Selectivity)**

Feed	C <sub>2</sub> <sup>=</sup>	C <sub>3</sub> <sup>=</sup>	C <sub>5</sub> <sup>+</sup>
1-butene	3.6%	22.7%	7.4%
MeOH	51.1%	34.2%	1.2%
MeOH + 1-butene	38.6%	35.0%	2.3%

10. It would be expected that the yields of a combined feed of a mixture of approximately a 1:1 MeOH and butene can be mathematically apportioned by their respective proportion in the conversion reaction. Thus, the *expected* yields of the combined reaction (MeOH + 1-butene) based on the individual component reactions are calculated by dividing the sum of the yields of a given product, e.g., ethylene, for the component reactions by the total conversion for both component reactions (34.2 + 100.0 = 134.2%) and multiplying the quotient by the percent conversion of the combined reaction (84.3%), as follows:

**Expected Yields of Combined Reaction Based on Yields of Component Reactions  
(Calculated From Actual Yield of Component Reactions and Percent Conversion)**

Feed	C <sub>2</sub> <sup>=</sup>	C <sub>3</sub> <sup>=</sup>	C <sub>5</sub> <sup>+</sup>
MeOH + 1-butene	34.3%	35.7%	5.4%

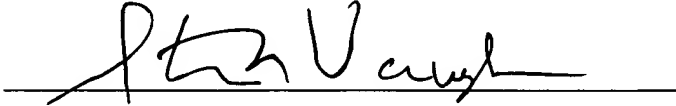
11. However, my experimental results, which are disclosed in the third row of the Table in Example 1 of the '399 Application, indicate that 45.72% of the product of the combined reaction is ethylene and 41.46% of the product is propylene. Accordingly, the actual ethylene yield of the combined reaction is 38.6% and the actual propylene yield is 35.0%. Thus, substantially more ethylene is produced by the combined reaction (4.3%) than would be expected, in view of the Kaiser patent teaching. An increase in ethylene yield of this magnitude is a highly desirable process improvement representing substantial commercial savings.

12. The above data shows that less propylene is produced by the combined reaction than would be expected (0.7%) in view of the teaching of the Kaiser patent. Additionally, based on the  $C_5^+$  yield of the component reactions, one of ordinary skill in the art would expect a  $C_5^+$  yield in the combined reaction of 5.4%. However, a  $C_5^+$  yield of 2.3% was actually observed, as indicated above. The  $C_5^+$  represent an unwanted byproduct and the present invention reduces the amount of  $C_5^+$  by more than half the expected amount. Reducing more than half of an undesirable byproduct alone is a significant commercial improvement.

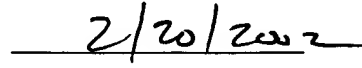
13. Based on the Kaiser patent disclosure and the component reaction data in my studies, one of ordinary skill in the art would expect to see a higher propylene yield in the combined reaction than was observed. In fact, a considerable increase in ethylene yield and slight decrease in propylene yield was observed in the combined reaction over the expected ethylene and propylene yields calculated from the component reactions. Additionally, a substantial 57% decrease in  $C_5^+$  yield was realized in the combined reaction over the expected yield based on the component reactions. These results would not be expected by a person of ordinary skill in the art when considering the Kaiser reference.

14. Based on my conclusion that when a mixed 1-butene/methanol feed stream contacts SAPO-34, there is a surprising increase in the amount of  $C_2^-$  produced and the surprising decrease in the selectivity of  $C_5^+$ , it is reasonable to expect that other small pore molecular sieve catalysts would behave in a similar manner.

15. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issue thereon.

A handwritten signature in black ink, appearing to read "Stephen N. Vaughn", written over a horizontal line.

Stephen N. Vaughn

A handwritten date "2/20/2002" in black ink, written over a horizontal line.

Date

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